



Responses of forest ecosystems in Europe to decreasing nitrogen deposition[☆]

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ABSTRACT

Average nitrogen (N) deposition across Europe has declined since the 1990s. This resulted in decreased N inputs to forest ecosystems especially in Central and Western Europe where deposition levels are highest. While the impact of atmospheric N deposition on forests has been receiving much attention for decades, ecosystem responses to the decline in N inputs received less attention. Here, we review observational studies reporting on trends in a number of indicators: soil acidification and eutrophication, understory vegetation, tree nutrition (foliar element concentrations) as well as tree vitality and growth in response to decreasing N deposition across Europe. Ecosystem responses varied with limited decrease in soil solution nitrate concentrations and potentially also foliar N concentrations. There was no large-scale response in understory vegetation, tree growth, or vitality. Experimental studies support the observation of a more distinct reaction of soil solution and foliar element concentrations to changes in N supply compared to the three other parameters. According to the most likely scenarios, further decrease of N deposition will be limited. We hypothesize that this expected decline will not cause major responses of the parameters analysed in this study. Instead, future changes might be more strongly controlled by the development of N pools accumulated within forest soils, affected by climate change and forest management.

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1. Introduction

Anthropogenic emissions drastically altered the global nitrogen (N) cycle (Fowler et al., 2013; Galloway et al., 2003; Vitousek et al., 1997), with human activities becoming the dominant contribution to the annual release of reactive N to the atmosphere (Fowler et al., 2015; Galloway et al., 2004). The increase in anthropogenic emissions arose from accelerated fossil fuel burning since the industrial revolution, the advent of the Haber-Bosch process to create reactive N from inert atmospheric N₂ at the start of the 20th century as well as an increase in mass transportation and livestock numbers

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(Engardt et al., 2017; Erisman et al., 2011). Today, 18% of the global anthropogenic nitrogen fixation can be attributed to combustion processes, 55% to fertilizer production, and 27% to biological N fixation in agriculture (Fowler et al., 2015). These activities have created benefits, such as the support of human nutrition by mineral fertilizers (Erisman et al., 2008). On the other hand, the release of reactive N causes considerable damages to human health (Van Grinsven et al., 2013) and induces changes in natural and semi-natural ecosystems, such as N deposition being one of the greatest threats to global plant diversity (Bobbink et al., 2010; Brink et al., 2011; Clark et al., 2013; Erisman et al., 2008; Soons et al., 2017; Vitousek et al., 1997).

In Europe N emissions and corresponding deposition increased from pre-industrial times till the mid-1980s, followed by a decrease since the 1990s (Engardt et al., 2017). The decline in N emissions is due to a combination of emission abatement policies and economic transformation (Erisman et al., 2003). In Europe's forests, N deposition caused a variety of changes, including impacts on tree productivity (De Vries et al., 2017b, 2006; Kahle, 2008), tree nutrition reflected in foliar concentrations (Jonard et al., 2015; Sardans et al., 2016b; Waldner et al., 2015), sensitivity of trees to biotic and abiotic stress (Bobbink and Hettelingh, 2011), the composition of understorey vegetation (Dirnböck et al., 2014; van Dobben and De Vries, 2017) and ectomycorrhizal fungal communities (van der Linde et al., 2018), to soil chemistry, and increased leaching of N from forest soils to surface and ground waters (Dise et al., 2009; Gundersen et al., 2006). In recent decades, much discussion took place to identify the mechanisms as well as the time frame by which forest ecosystems are impacted by elevated nitrogen deposition. The concept of *nitrogen saturation* (Aber et al., 1998, 1989; Ågren and Bosatta, 1988; De Vries and Schulte-Uebbing, 2018; Lovett and Goodale, 2011) suggests a set of reactions including loss of plant species diversity, N losses with seepage water, soil acidification, and growth reduction. A recent perspective on the stages of N saturation is depicted in Fig. 1. Ecological understanding is used

to determine critical loads of N deposition defined as 'a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge' (Nilsson and Grennfelt, 1988). Critical loads underpin emission protocols at the European scale such as the Revised National Emissions Ceilings Directive (NECD) and are also applied for example in North America (Pardo et al., 2011; Schindler and Lee, 2010) and Asia (Duan et al., 2016). Exceedances of critical loads indicate risks of adverse effects on various aspects of forests, such as tree nutrition and forest biodiversity (De Vries et al., 2015; Nordin et al., 2005; Waldner et al., 2015).

A large part of the ecological research in this context focused on the responses of forest ecosystems to elevated N deposition resulting in N saturation or the exceedance of critical loads. However, much less attention was paid to the potential dynamics of a "recovery" from high N loads although a decline of N deposition to Europe can be observed since the 1990s. The average deposition of inorganic N across all land-use types in Europe decreased from 10.3 kg N ha⁻¹ a⁻¹ in 1990 to 6.6 kg N ha⁻¹ a⁻¹ in 2018 (after Engardt et al. (2017), data kindly provided by Magnuz Engardt and David Simpson). The trends are distributed heterogeneously in space. While many forests in areas with higher absolute levels of N deposition (e.g. in Central and Western Europe) experienced a decrease in N inputs, less clear trends have been reported for Northern Scandinavia and parts of Southern Europe (Figs. 2 and 3). Note that despite these reductions, 62% of the European ecosystem area was at risk of eutrophication due to the exceedance of the critical load for eutrophication in 2015 (Slootweg et al., 2015).

This study addresses the response of European forest ecosystems to decreasing N deposition. We review published results from observational and experimental studies on well-monitored parameters: soil acidification and eutrophication, foliar chemistry, ground vegetation composition, tree vitality, and tree growth. This set of indicators covers a range between *endpoint metrics*, i.e. aspects

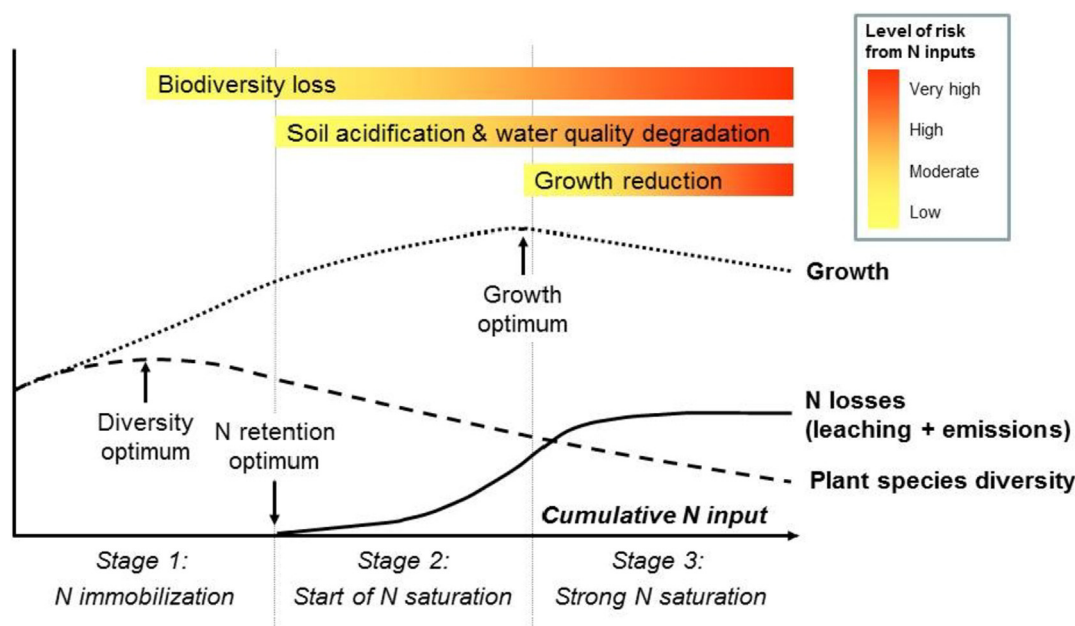


Fig. 1. Hypothetical relationship between the stage of nitrogen saturation and the effects on terrestrial ecosystems in terms of soil processes, vegetation changes and growth. This figure is an update of the figure by Aber et al. (1998) (after De Vries and Schulte-Uebbing (2018)). It illustrates the trade-off between the initial positive impact of nitrogen enrichment on tree growth and related carbon sequestration on the one hand and the negative impact on ecosystem services (e.g. water quality regulation by nitrogen retention) and on biodiversity on the other hand.

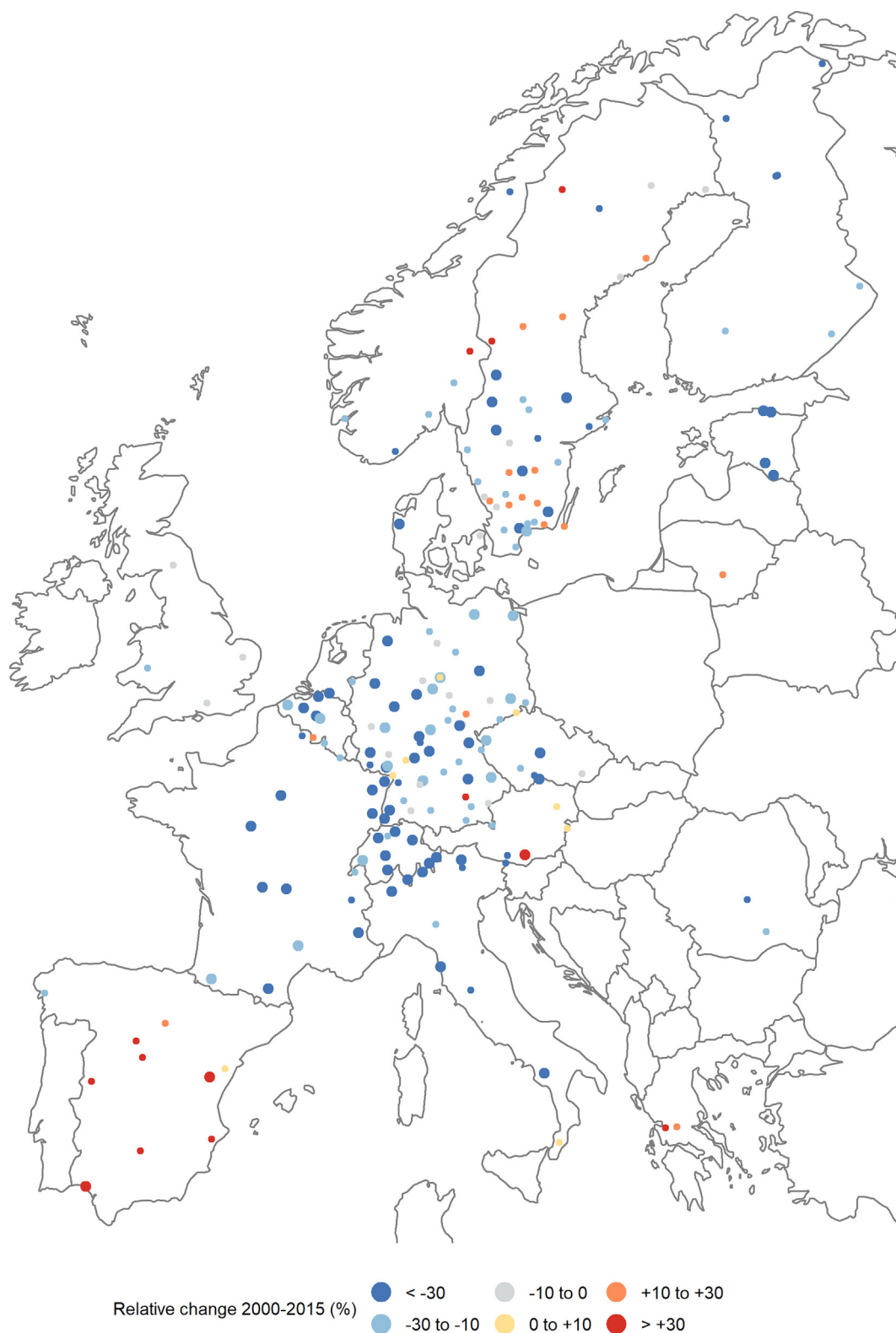


Fig. 2. Relative change of throughfall deposition of inorganic nitrogen at the intensive monitoring sites of the UNECE ICP Forests programme network between 2000 and 2015 (redrawn after Schmitz et al., 2018). Large dots indicate statistically significant trends; trends represented by small dots are not statistically significant.

of the environment that are directly relevant to people (e.g. tree growth) and *midpoint metrics*, i.e. parameters that are well-suited to measure progress towards desired environmental states (e.g. plant tissue concentrations) (Rowe et al., 2017). While results are

limited to Europe, references have also been included relating to observations and experiments in the United States (US). For a detailed overview of impacts of decreased N deposition in the US, we refer to Gilliam et al. (2018, in press).

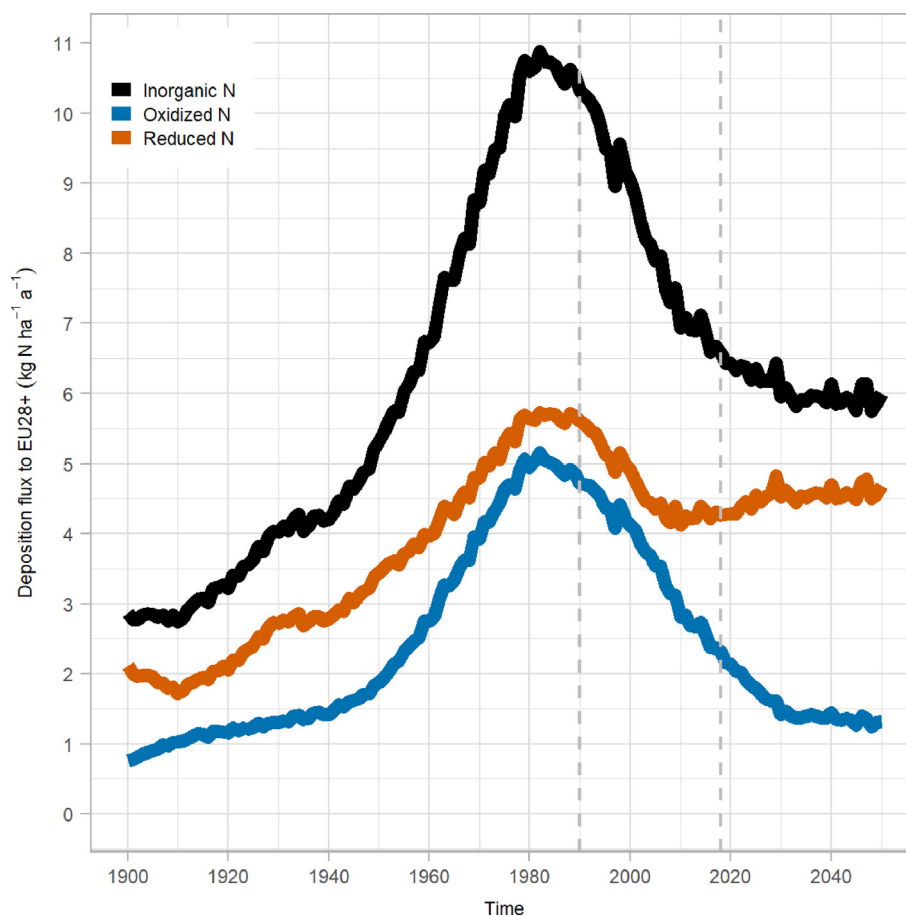


Fig. 3. Average deposition of oxidized, reduced and total N between 1900 and 2050 to the EU28, Norway and Switzerland according to EMEP model results (after Engardt et al. (2017), data kindly provided by Magnuz Engardt and David Simpson). Vertical dashed lines indicate the years 1990 and 2018. Future reductions are expected to be small and inorganic N deposition is likely converging to a level approximately twice as high compared to 1900.

2. Soil acidification and eutrophication

Atmospheric deposition of reactive nitrogen compounds such as nitrate (NO_3^-) and ammonium (NH_4^+) contributes to acidification and eutrophication of forest soils (Driscoll et al., 2006). Soil acidification involves accelerated losses of mineral nutrients (base cations, i.e. Ca^{2+} , K^+ and Mg^{2+}) and potential for the mobilization of toxic aluminium (Al), both of which can compromise tree health (Driscoll et al., 2006; Boudot et al., 1994; De Vries et al., 2014; De Wit et al., 2010). N deposition contributes to elevated soil solution NO_3^- concentrations and soil N stocks (Driscoll et al., 2001). This enrichment can have a variety of effects on trees and ground vegetation, covered in the subsequent chapters. NO_3^- concentrations in soil solution are a good indicator for the soil N status. Important determinants of NO_3^- leaching are the C/N ratio of the forest floor (Gundersen et al., 1998a) and N deposition rates (Dise and Wright, 1995), as well as a variety of other site and stand characteristics controlling the ecosystem N cycling (Lovett and Goodale, 2011). Generally, elevated NO_3^- concentrations in soil solution are an indication of N availability in excess of biotic demand. Spatial patterns of soil solution NO_3^- are highly variable but partly reflect spatial patterns in N deposition, with higher levels in the Netherlands, Belgium, parts of Germany, Switzerland, and Denmark and lower levels in parts of France, Norway, Northern Sweden, and Finland (Boxman et al., 2008; De Vries et al., 2007; Evans et al., 2001; Gundersen et al., 1998a; Jonard et al., 2012; Mellert et al., 2008; Moffat et al., 2002; Pannatier et al., 2010; Pihl Karlsson

et al., 2011; Rothe et al., 2002; Ukonmaanaho et al., 2014; van der Heijden et al., 2011; Verstraeten et al., 2012). There are relatively fewer reports of elevated NO_3^- in soil solution in Southern and Eastern Europe, and N deposition is mostly lower in these regions (Waldner et al., 2014).

Observational studies

At the European scale, studies examining trends in soil solution N show weak or non-significant trends. For example, Johnson et al. (2018) found a weakly significant ($p < 0.1$) reduction in NO_3^- concentrations at 40–80 cm depth corresponding to a decrease of 30% over 10 years when analysing data from 162 plots across Europe between 1995 and 2012. They found no significant trend in 10–20 cm depth. An earlier analysis (from the early 1990s to 2006) using a similar dataset found mostly non-significant trends in soil solution inorganic N concentrations (Iost et al., 2012). These studies did not focus specifically on areas with high N deposition and included many sites from N limited areas of Northern Europe. Within Europe, national and regional studies show variable results. For example, in the Netherlands and Flanders soil solution NO_3^- declined in response to decreasing N deposition (Boxman et al., 2008; Verstraeten et al., 2012). In contrast, an intensive study at the site Solling in Germany found NO_3^- continued leaching from a spruce (*Picea abies*) stand and increased at a beech (*Fagus sylvatica*) stand despite decreasing N deposition between 1973 and 2013, indicating a reduction of the N retention capacity of the soil over

Table 1
Summary of trends in soil solution chemical characteristics indicative for eutrophication and acidification status (concentration of NO_3^- , base cations (BC, i.e. Ca^{2+} , K^+ and Mg^{2+}) and total aluminium (Al_{tot}), pH, equivalent ratio of BC and Al_{tot} (BC: Al_{tot}), acid-neutralizing capacity (ANC) and ionic strength) from studies across Europe.

Effect	Trend				
	↑	↑/↔	↔	↔/↓	↓
NO_3^-		Meesenburg et al. (2016) (Germany)	Johnson et al. (2013) (Ireland), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden), Vanguelova et al. (2010) (UK)	Pannatier et al. (2010) (Switzerland), Sawicka et al. (2016) (UK), Ukonmaanaho et al. (2014) (Finland)	Boxman et al. (2008) (Netherlands), Oulehle et al. (2011) (Czech Republic), Verstraeten et al. (2012), Verstraeten et al. (2017) (Flanders)
pH	Akselsson et al. (2013), Löfgren et al. (2011) (Sweden), Verstraeten et al. (2016) (Flanders)	Vanguelova et al. (2010), Sawicka et al. (2016) (UK), Fölster et al. (2003), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden), Johnson et al. (2013) (Ireland)			Boxman et al. (2008) (Netherlands), Jonard et al. (2012) (Wallonia)
BC			Vanguelova et al. (2010) (UK), Johnson et al. (2013) (Ireland)	Graf Pannatier et al. (2011) (Switzerland), Sawicka et al. (2016) (UK)	Jonard et al. (2012) (Wallonia), Verstraeten et al. (2012) (Flanders), Boxman et al. (2008) (Netherlands), Fölster et al. (2003), Akselsson et al. (2013) (Sweden)
Al_{tot}	Jonard et al. (2012) (Wallonia), Fölster et al. (2003) (Sweden)		Sawicka et al. (2016) (UK)	Vanguelova et al. (2010), Löfgren et al. (2011), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden), Johnson et al. (2013) (Ireland)	Verstraeten et al. (2012) (Flanders), Boxman et al. (2008) (Netherlands)
BC: Al_{tot}		Meesenburg et al. (2016) (Germany)		Graf Pannatier et al. (2011) (Switzerland)	Verstraeten et al. (2012) (Flanders)
ANC	Akselsson et al. (2013), Löfgren et al. (2011) (Sweden), Verstraeten et al. (2012) (Flanders)	Fölster et al. (2003), Löfgren and Zetterberg (2011), Pihl Karlsson et al. (2011) (Sweden)			
Ionic strength				Löfgren and Zetterberg (2011) (Sweden)	Löfgren et al. (2011) (Sweden), Verstraeten et al. (2012) (Flanders), Vanguelova et al. (2010) (UK)

time (Meesenburg et al., 2016). Other studies found no trends in NO_3^- soil solution concentrations during periods of stable N deposition (e.g. Alewell et al., 2000.; Johnson et al., 2013; Pannatier et al., 2010). At a heavily acidified forest in the Czech Republic, NO_3^- concentrations in soil solution declined despite no decrease in N deposition. This was due to an increase in N uptake by vegetation and changes in organic matter cycling as the soil became less acidic (Oulehle et al., 2011). Where soil solution NO_3^- decreased, it is generally accompanied by a decrease in base cations and total Al concentrations, while soil solution pH and acid neutralizing capacity (ANC) showed no uniform trends in recent decades (Iost et al., 2012; Johnson et al., 2018). Recovery from acidification primarily occurs on poorly buffered, acidic soils while acidification progresses on better buffered soils despite large decreases in sulphur (S), and to a lesser degree, N deposition (Johnson et al., 2018). The absence of a uniform recovery of soil solution from acidification agrees with trends in bulk soil chemistry. Cools and De Vos (2011) found that base saturation increased in soils with low buffering capacity but decreased in soils with initially higher base saturation across Europe. A similar result was found for the Netherlands between 1990 and 2015 (De Vries et al., 2017a). Table 1 summarizes results on trends of soil solution eutrophication and acidification status from studies across Europe.

Experimental studies

In addition to observational studies, also field experiments provide information on changes of the soil chemical status under decreasing N deposition. In this context, the NITREX and EXMAN nitrogen manipulation experiments at several sites in Europe are a valuable source of information (Wright and Rasmussen, 1998). At three NITREX sites, throughfall N deposition was decreased from 36 to 50 kg N ha⁻¹ a⁻¹ to 5–16 kg N ha⁻¹ a⁻¹ by roofing. A decline in N leaching became apparent within the first three years of treatment at all three sites (Beier et al., 1998; Boxman et al., 1998; Emmett et al., 1998; Gundersen et al., 1998b). A similarly fast response in N leaching has been observed from a roofing experiment in southern Norway (Wright et al., 1993). These results indicate that continuous high N inputs are required to sustain N leaching in most forest ecosystems, suggesting that decreasing deposition quickly translates into improvements in soil water quality (Emmett et al., 1998). This, however, also implies that considerable amounts of N deposited over the last decades are retained and that the return of the ecosystem to the original N status is potentially slow (Gundersen et al., 1998b). In contrast to these findings, also unchanged or increased N leaching despite decreased deposition was occasionally reported from observational (Meesenburg et al., 2016) and experimental studies (Emmett et al., 1998).

Summary

Long-term monitoring data provides information on NO_3^- concentrations in soil solution as an indicator for the soil N status. Despite considerable heterogeneity, indications for a decreasing trend in soil solution NO_3^- concentrations at the European scale exist. Experimental studies tend to report a faster and more pronounced reaction of soil solution NO_3^- concentrations compared to the findings from large-scale observational studies. In the experiments the magnitude and speed of decrease in N supply was larger compared to trends in N deposition in most parts of Europe. Furthermore, longer-term changes in soil microbial activity (e.g. mineralization rates) might be reflected to a larger degree in the observational studies compared to experimental studies which often focus on the time period immediately after the onset of the artificial decrease of N supply. Nevertheless, both types of studies suggest indications of a response in soil solution NO_3^- concentrations to decreases in N deposition. Soil acidification shows non-uniform tendencies across Europe despite large-scale decreases in N, and especially S, deposition.

3. Understory vegetation

Forests provide habitat for understory vegetation, bryophytes, lichens as well as microbial and animal communities. While N is a limiting resource for many organisms (Vitousek and Howarth, 1991), the efficiency with which it is used is species-specific (Chapin, 1980). As a consequence, more N causes some species to thrive on the expense of others, usually causing a net loss in species diversity (Suding et al., 2005). Besides this effect on interspecific competition, changes in N deposition can also modify herbivory, interactions with fungi, and invasibility by exotic species, thereby affecting understory species composition (Gilliam, 2006). In managed forests these mechanisms are rarely reflected in the composition of the main tree species for they are typically intentionally chosen by forest managers. In contrast, forest understory vegetation, bryophytes, lichens, mycorrhiza, and soil fauna can be expected to be affected by N availability in addition to other environmental factors such as light availability, temperature, moisture, and nutrients other than N. The responses of these groups to elevated N deposition encompass changes in the abundance of species, alteration in the identity of species (species composition), and pauperization of local and regional species diversity (Bobbink et al., 2010; Farrer and Suding, 2016; Hautier et al., 2009; Nijssen et al., 2017). Fig. 4 exemplifies effects of N deposition on forest understory vegetation for lichen diversity and herb layer plant community composition.

Observational studies

While there are several observational studies on the reaction of forest understory diversity to elevated N deposition, to our knowledge, none of them focused specifically on the response to declining N deposition. These studies confirm an increase in nitrophilic forest understory plant species on the expense of oligophilic species both in European-wide (Dirnböck et al., 2014; van Dobben and De Vries, 2017) as well as regional contexts (Bobbink and Hettelingh, 2011 and references therein; Heinrichs and Schmidt, 2016; Keith et al., 2009; Roth et al., 2015). Besides N deposition, litter quality, light availability, density of large herbivores, and differences in forest management were also important drivers of change in understory plant communities (Bernhardt-Römermann et al., 2015; Perring et al., 2017; Verheyen et al., 2012). These changes in species composition do not, however, seem to be accompanied by a broad scale, synchronized decline in

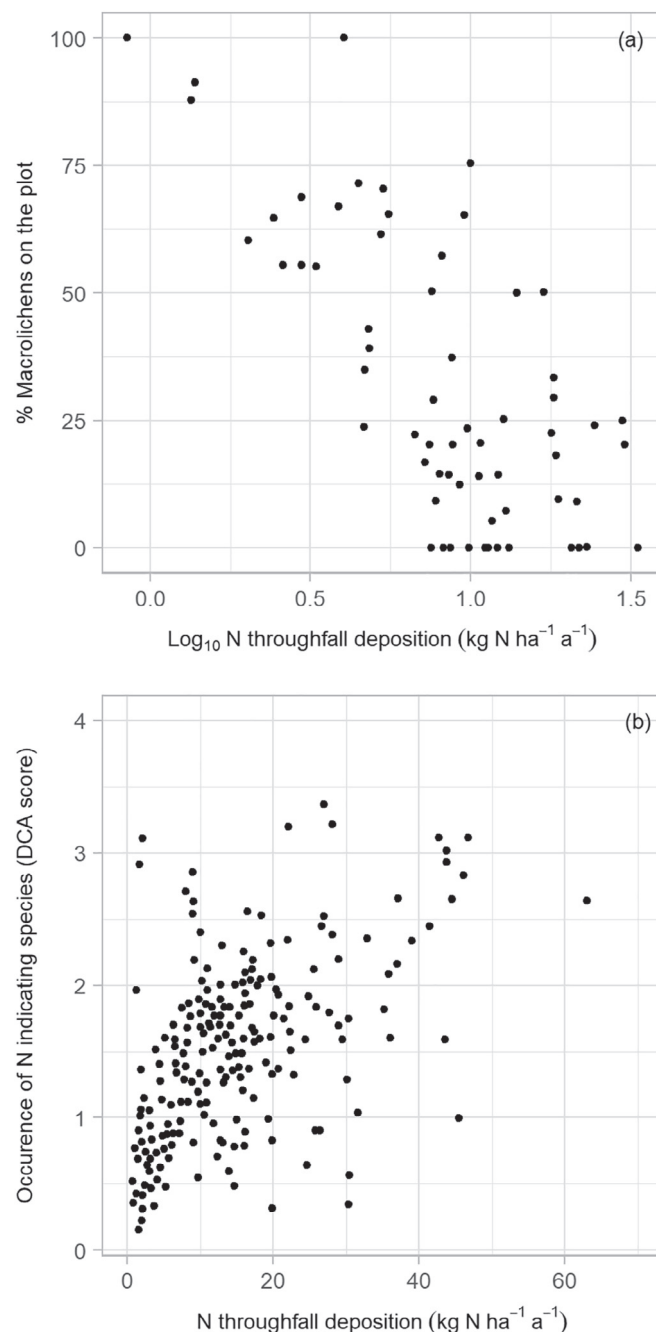


Fig. 4. Examples for the effects of N deposition on forest understory vegetation. (a) Relationship between lichen diversity (proportion of macrolichen species among all lichen species) and N throughfall deposition based on 83 forest plots across Europe. Reprinted from Giordani et al. (2014) with permission from Elsevier. (b) Relationship between the occurrence of nitrogen indicating species and N throughfall deposition based on a detrended correspondence analysis (DCA) of the floristic composition of the herb layer at 488 forest plots in the nemoral zone of Europe. Scores on the fourth axis of the DCA are positively correlated with nitrogen deposition. Redrawn from Seidling et al. (2008) by permission of the publisher (Taylor & Francis Ltd, <http://www.tandfonline.com>).

plant diversity in European forests (Dirnböck et al., 2014; van Dobben and De Vries, 2017; Verheyen et al., 2012).

In contrast, elevated N deposition has clearly contributed to a dramatic diversity loss in epiphytic lichens in many European forests (Bobbink and Hettelingh, 2011; Giordani et al., 2014; Hauck et al., 2013; Mayer et al., 2013). Similarly, major impacts in the

community composition and diversity of mycorrhiza were identified at the European level (Suz et al., 2014; van der Linde et al., 2018) and in various regional studies (Bobbink and Hettelingh, 2011, references therein). Furthermore, diversity effects of N deposition on one receptor can indirectly affect others such as soil fauna and mammals because effects cascade from e.g. plants to animal species (Nijssen et al., 2017) or from soil microbes to plants (Farrer and Suding, 2016). However, studies detailing the link between N deposition and animal diversity in Europe's forests are scarce, partly due to the complex dynamics of animal populations and corresponding food-webs (Nijssen et al., 2017).

Experimental studies

In addition to these findings from observational studies, a limited number of N manipulation experiments report on changes in understory vegetation in response to decreasing N input. Strengbom et al. (2001) compared vascular plant, fungi, and bryophyte communities between control and treatment plots at two experimental forest sites in Sweden where N fertilization was cancelled nine and 47 years prior to the analyses, respectively. They found differences in the vascular plant community at the site where treatment ended nine years ago but no longer at the site where treatment was cancelled 47 years ago. Nevertheless, the fungi and bryophyte communities deviated from the control plots at both sites. Sujetovienė and Stakėnas (2007) report on changes in pine forest understory plant community in response to drastic emission reductions from a close-by fertilizer plant in Lithuania. They found a decrease in nitrophilous species within the 16 years between two ground vegetation studies (1988 and 2004). It should be noted that also the light conditions and the acidity status of the respective forest stands changed over the same time. In one of the NITREX experiments, N-indicating fern cover significantly decreased after 5 years of reduction of N deposition from 60 kg N ha⁻¹ a⁻¹ to 5 kg N ha⁻¹ a⁻¹ by roofing. A recovery of other species was not recorded (Boxman et al., 1998).

Also findings from grassland vegetation experiments might be informative for the question of forest understory vegetation response to decreasing N deposition. Stevens et al. (2012) found significant differences in Ellenberg N values between control and treatment plots 15 years after cessation of N fertilization in mesotrophic grassland in the UK. Shi et al. (2014) report on the vegetation composition three years after cessation of N fertilization at a sandy grassland site in Northeast China. They found that the vegetation at the control and the formerly treated plots still differed although indications for an ongoing process of recovery were apparent. Storkey et al. (2015) report that grassland biodiversity largely recovered over a period of 20 years of decreasing ambient N deposition, based on observations from the control plot of a fertilization experiment in the UK. The pronounced recovery was potentially supported by the regular export of N from the ecosystem by haying (Tilman and Isbell, 2015).

Summary

Recent studies based on large-scale monitoring data find shifts in understory community composition in response to high levels of N deposition but do not report on responses to decreasing N deposition. Results from experimental studies suggest that while the recovery of understory vegetation from high N inputs is possible, time-lags in the order of decades are to be expected. One mechanism causing these delays is that in regions where high N deposition eradicated source populations, back-colonization will be particularly difficult (Clark and Tilman, 2010; Dullinger et al., 2015). The complex consequences of such effects have already been

shown for land management legacies' impact on dispersal dynamics and subsequent community alterations (e.g. Burton et al., 2011). Strong recovery delay due to dispersal limitation can be expected for epiphytic lichens because regional species extinctions were particularly pronounced (Hauck et al., 2013). We hypothesize that this delay in the response of understory vegetation to decreases in N deposition partly explains the absence of corresponding trends in Europe-scale observational studies. In addition, changes in other environmental conditions like light availability, forest management, sulphur deposition, habitat loss and fragmentation, climate impact, and non-native species invasion (see e.g. Perring et al., 2017) superimpose on the signal of N deposition in forest understory communities.

4. Tree nutrition

Foliar element concentrations and their ratios reflect the nutritional status of trees. Unbalanced N:P ratios in foliar tissues are frequently associated with defoliation (Bontemps et al., 2011; Ferretti et al., 2015; Veresoglou et al., 2014; Waldner et al., 2015) and an increasing risk of attacks by parasites (Flückiger and Braun, 1998) and herbivores (Pöyry et al., 2016) as well as decreasing plant capacity to respond to abiotic stressors such as drought, warming, and frost (Fangmeier et al., 1994; Sardans and Peñuelas, 2012). Furthermore, changes in N:P ratio in foliar tissues can have several consequences in forest trophic chains (Peñuelas et al., 2013). For example, increases in foliar-litter N:P ratios have been associated with shifts in community composition and decreases in species richness in soil communities and understory vegetation in some European forests (Peñuelas et al., 2013). Unbalanced plant N:P ratios can reduce the resistance to biotic stressors such as the competition against invasive species (Sardans et al., 2016a).

Observational studies

The status and trends of tree nutrition are highly variable across Europe. At the European scale, two recent studies report tendencies of decreasing foliar N concentrations for beech and oak, covering the periods 1992–2009 and 2000–2015, respectively (Jonard et al., 2015; Sanders et al., 2017b). To a lesser extent, decreases are also indicated for spruce, while stable or slight increasing foliar N concentrations are reported for pine (*Pinus sylvestris*). At the same time, however, the mass per needle/leaf significantly increased for spruce and beech, causing an overall increase in the N content per needle/leaf despite the decreasing concentrations ("dilution effect", Jonard et al., 2015). At the local or regional level, studies based on data from 1990 onward report stable N concentrations or moderate changes in both directions (Jonard et al., 2012; Verstraeten et al., 2017; Wellbrock et al., 2016). Analysis restricted to, or including data from before 1990 frequently (Duquesnay et al., 2000; Hippeli and Branse, 1992; Mellert et al., 2004 for pine; Prietzel et al., 1997; Sauter, 1991) but not always (Braun et al., 2010; Mellert et al., 2004) report increasing foliar N concentrations or contents across Europe. Although not focused on temporal trends, other studies suggest a general effect of N deposition on foliar N concentrations based on analyses of large-scale spatial data (De Vries et al., 2003; Sardans et al., 2016b).

Foliar P concentrations decreased continuously according to studies analyzing data from 1990 onward in the important forest species in central and northern Europe, such as pine, spruce, beech, and sessile oak (*Quercus petraea*), resulting in low or deficient foliar P status on 22%–74% of the plots depending on tree species (Ferretti et al., 2015; Jonard et al., 2015, 2012; Talkner et al., 2015). For N:P, increasing ratios have been observed in several studies at European scale based on data after 1990 (Jonard et al., 2015; Sanders et al.,

2017a; Talkner et al., 2015). Apart from N:P imbalances, also trends towards increasing N:K and N:S ratios have been observed in a Europe-wide study while the N:Mg ratio was decreasing (Jonard et al., 2015). N deposition can cause deficiencies in other nutrients than N and nutrient imbalances due to a range of effects, including stimulation of plant growth (dilution effect) and negative effects on tree nutrient acquisition by modifying mycorrhizal associations (De Witte et al., 2017; Jonard et al., 2015; Peñuelas et al., 2013; Sardans et al., 2016b). Thus, the decreasing tendencies in foliar concentrations of nutrients other than N and increasing N:other element ratios suggest that N availability is still high in many regions across Europe and do not imply a recovery from high N deposition yet.

Experimental studies

Besides observational studies, a number of experiments provide indication on the reaction of foliar element concentrations to decreased N supply. In one of the abovementioned NITREX roofing experiments, a decrease in needle N concentrations and an improvement (reduction) of the N:Mg and N:K ratio is documented after three years (Boxman et al., 1998). At the other two sites, no significant reductions in foliar N concentrations were observed six years after the treatment started (Emmett et al., 1998). Högborg et al. (2006) report average foliar element concentrations for the time period seven to twelve years after the cessation of an N addition treatment. Foliar N concentration clearly decreased and other elements showed minor increases. Twenty years after termination of the N fertilization at the same site, foliar N concentrations were still slightly elevated compared to the control (Högborg et al., 2014). Similarly, Blaško et al. (2013) report a recovery (decrease) of foliar N concentrations based on measurements 17 and 19 years after the termination of a N fertilization experiment, while also still slightly exceeding the levels at the control plot. Results from grassland and moorland fertilization experiments report that foliar N concentrations had decreased when measured 7–15 years after cessation of the N addition (Clark et al., 2009; Edmondson et al., 2013; Stevens et al., 2012). These findings from experiments indicate that decreases in N deposition can be expected to be reflected in foliar N concentrations with a lag time of a several years.

Summary

Despite the large heterogeneity in trends in tree nutrition, studies based on large-scale long-term monitoring data report tendencies of decreasing foliar N concentrations for beech, oak and to a lesser extent for spruce. The degree to which the decreasing trends in N deposition contribute to these trends is not clear. On the one hand, decreasing tendencies of NO_3^- concentration in soil solution (see “Soil acidification and eutrophication”), findings from experimental studies as well as large-scale studies documenting the relation between spatial patterns of N deposition and foliar N concentrations suggest that the decrease in N deposition could have affected foliar N concentrations. On the other hand, the cutback in N deposition across Europe is typically far smaller compared to experimental treatments and might not yet have led to a widespread decrease in N availability for tree nutrition in a relevant magnitude (Braun et al., 2010; Mellert et al., 2017; Riek et al., 2016). The increase in foliar mass (dilution effect, Jonard et al., 2015) likely explains a considerable proportion of the decrease in foliar N concentrations. Furthermore, decreasing tendencies in other elements and increasing N:other element ratios do not indicate recovery from high N availability. Further analyses are required to gain a better understanding where and to what extent

changes in N deposition or other mechanisms control tree nutrition across Europe and which time lags are involved.

5. Tree vitality

The concept of “vitality” of forests is linked to several inter-related aspects, including above- and below-ground growth, tree nutrition as well as the susceptibility of trees to biotic (e.g. insects) and abiotic (e.g. climatic extremes) stress. Tree crown condition is often interpreted as an aggregated measure of tree vitality because it reflects the impacts of several different environmental drivers. It is typically measured in the form of the degree of ‘crown defoliation’ (Eichhorn et al., 2016).

Observational studies

Several studies addressed the link between nitrogen deposition and defoliation at the European scale (e.g. Ferretti et al., 2015; Klap et al., 2000) but to our knowledge none reports explicitly on the effect of decreased N deposition. Existing studies focus on the relative importance of air pollution among other determinants of crown condition like climate, soil, and stand age. The results reflect the complexity and spatial heterogeneity of the underlying processes. For example, Ferretti et al. (2015) found that N-related variables improved defoliation models based on data from 71 plots across Europe. Higher N deposition led to higher percentage of defoliated trees for beech and spruce, while the effect was opposite for pine. Similarly, Vitale et al. (2014) and De Marco et al. (2014) found aspects of N deposition to be relevant determinants of crown condition for several species across Europe, with varying direction of the effect. Other studies found weak or no relation between defoliation and N deposition (Hendriks et al., 2000; Klap et al., 2000; Solberg and Tørseth, 1997; Staszewski et al., 2012). In a regional study, Armolaitis and Stakenas (2001) report on the response of the crown condition of a pine forest to emission reductions from a close-by fertilizer plant in Lithuania. Refoliation began 6–7 years after the decrease of air pollution.

N-induced effects on vitality

The mechanisms by which excess N supply can cause a net decrease in tree vitality can be complex, interlinked and only episodically apparent, including increased susceptibility to insect attacks, pathogens, frost and storm damages (Bobbink and Hettelingh, 2011), changes in mycorrhiza (Arnolds, 1991; Braun et al., 2010; De Witte et al., 2017; Duquesnay et al., 2000; Jaenike, 1991; van der Linde et al., 2018), changes in the rooting system and aluminum toxicity to roots (Dziedek et al., 2017; Godbold and Kettner, 1991; Haynes, 1982; Jonard et al., 2012; Ostonen et al., 2007), depletion of base cations due to NO_3^- leaching (Jonard et al., 2012; Prietzel et al., 1997) or problematic P supply (Jonard et al., 2015; Mellert and Ewald, 2014; Neiryneck et al., 1998; Ochoa-Hueso et al., 2013; Peñuelas et al., 2013; Sardans et al., 2015; Sardans and Peñuelas, 2012; Thelin et al., 1998). Tree species, stand age, soil, and meteorological conditions as well as other local factors co-determine these symptoms.

Summary

Tree crown condition provides an aggregated measure of tree vitality. Studies evaluating spatial and temporal patterns of crown condition based on long-term monitoring data come to different conclusions regarding the relative importance and direction of the effect of N deposition. To our knowledge, no large-scale response to decreasing N deposition has been reported. N deposition can have

both a positive (fertilizing) effect on tree vitality (crown condition) but also contribute to a range of adverse effects. We assume that the high complexity and spatio-temporal variability of these mechanisms is partly causing the difficulty to detect signals of decreasing N deposition in tree vitality. In addition, factors like site, stand age, drought, and frost can have strong effects on vitality independent of N deposition (e.g. Eickenscheidt et al., 2016; Klap et al., 2000).

6. Tree growth

Tree growth provides the primary economic benefit from most forest sites and is an important process in forest CO₂ budgets. Aber et al. (1998) hypothesized that net primary production of trees will show an increasing followed by a decreasing (unimodal) response with ongoing nitrogen saturation (comp. Fig. 1). The underlying assumption is that low to moderate levels of N deposition will relieve trees from growth limitation due to originally widespread N shortage (Aber et al., 1995; De Vries et al., 2009; Kahle, 2008; Schulte-Uebbing and De Vries, 2017; Solberg et al., 2009; Sutton et al., 2008; Vitousek and Howarth, 1991). However, when N deposition exceeds a certain level, the stimulating effects diminish due to the antagonistic effects applying to overall tree vitality (see above), e.g. of soil acidification, nutrient imbalances and increased susceptibility to biotic and abiotic stress (Aber et al., 1998; De Vries et al., 2014; Dobbertin, 2005). For example, beneficial effects for tree growth by recovery from acidification have been documented in Europe and the US (Mathias and Thomas, 2018; Juknys et al., 2014).

Observational studies

There are various broad-scale and regional studies investigating the effect of N deposition on tree growth, while accounting for the impacts of other drivers, such as changes in temperature and precipitation (e.g. Braun et al., 2017; Kint et al., 2012; Kolář et al., 2015; Solberg et al., 2009). In these studies, changes in growth patterns have rarely been explicitly linked to declining trends in nitrogen deposition. In some cases, a simultaneous decrease in S and N deposition complicated the separation of effects (Juknys et al., 2014; Nellesmann and Thomsen, 2001). However, the results of these studies provide indications for the threshold level of N deposition at which growth enhancement and growth reductions can be expected (Braun et al., 2017; Kint et al., 2012). For example, field monitoring data of forest growth at more than 300 plots in Europe suggest a non-linear growth response to N deposition between 3 and 60 kg N ha⁻¹yr⁻¹ with a threshold near 35 kg N ha⁻¹yr⁻¹ (Solberg et al., 2009). Kint et al. (2012) documented a non-linear growth response, in terms of basal area increment (BAI), to increasing N availability for 180 oak and beech plots in Flanders throughout the 20th century (the period 1901–2008). They found positive effects of N deposition on BAI up to 20–30 kg N ha⁻¹ yr⁻¹ and declining growth above these levels. Etzold et al. (2014) found a non-linear relation between NPP and N deposition, with the positive effect flattening off at sites with an N deposition above 20 kg N ha⁻¹yr⁻¹, based on data from intensive monitoring plots in Switzerland. In experimental and observational studies in forests in Switzerland, Flückiger et al. (2011) found a growth-stimulating effect of N which turned into no effect or a decrease of growth with increasing duration or magnitude of the N input. Anders et al. (2002, in Bobbink and Hettelingh, 2011) reported growth-reducing effects of N deposition on Scots pine stands in the north-east of the German Northern Lowland in the vicinity of N emission sources with deposition rates exceeding 35 kg N ha⁻¹ a⁻¹, while for other locations and tree species, accelerated growth was observed at open field deposition rates exceeding 10–15 kg N ha⁻¹ a⁻¹.

Experimental studies

Further information on the growth response of trees to different levels of N deposition originates from field experiments. For example, in one of the NITREX experiments, Boxman et al. (1998) report a significant increase in growth after three years of artificially decreasing N deposition rates by roofing. It should be noted, however, that in this experiment not only N but also S deposition decreased. Högborg et al. (2006) found that very high rates of N addition (90–180 kg N ha⁻¹ a⁻¹) led to an increase in tree growth only until a cumulative amount of approximately 1 t N ha⁻¹ while further N addition lowered the gain in wood volume. In a similar experimental setup, Blaško et al. (2013) observed that a strongly fertilized plot (90–180 kg N ha⁻¹ a⁻¹) had a lower long-term average productivity than other fertilization levels (30–120 kg N ha⁻¹ a⁻¹) but still more than the control plot. These results support the perspective that improved N supply has a positive effect on growth in case of N limitation and can act negatively in case of excess N (Flückiger et al., 2011).

Global meta-analyses also confirm thresholds in the growth response of trees to N deposition. For example, Tian et al. (2016) analysed a dataset of 44 experimental studies from wetland, grassland, temperate, and boreal forest (most data are from temperate forest). They found that the effect of N input on above-ground net primary production switches from increase to decrease at approximately 50–60 kg N ha⁻¹ a⁻¹. Schulte-Uebbing and de Vries (2017) found that the N-induced increase in carbon sequestration was significantly lower at higher ambient N deposition rates (above 15 kg N ha⁻¹ a⁻¹), reviewing results from forest fertilization experiments in temperate, boreal, and tropical forests. Field data of maximum rates of photosynthesis against N deposition for 80 forested plots over the world indicated an increase in photosynthesis up to an N deposition near 10–15 kg N ha⁻¹ a⁻¹ followed by no further change up to 35 kg N ha⁻¹ a⁻¹ (Fleischer et al., 2013).

Summary

We did not find an indication for a large-scale response in tree growth to decreasing N deposition. However, results from observational and experimental studies corroborate the concept of a unimodal response of tree growth to N deposition. Estimates of thresholds above which N deposition negatively affects tree growth range from as low as 15–20 kg N ha⁻¹ a⁻¹ to very high levels only relevant under experimental conditions. This suggests that particularly polluted forest stands (mostly located in Central and Western Europe) might have benefitted from declining N deposition, as decreases have been strongest in the formerly most polluted regions. Few trends of decreasing N deposition have been observed in less polluted areas like Northern Scandinavia, suggesting that a growth decline due to decreased N deposition in these areas is less likely.

7. Conclusion and outlook

Results from observational studies across Europe for responses in soil, ground vegetation, and trees (nutrition, growth and vitality) to decreasing N deposition indicate considerable spatial variability in the trends published for these parameters. For soil solution NO₃⁻ concentrations and potentially also for changes in foliar N concentrations, indications for a reaction to decreased nitrogen deposition exist. We found several studies reporting on the effects of N deposition on understory vegetation, tree growth or tree vitality, but none of them focused specifically on responses to declining N deposition. For tree growth, these studies suggest a positive effect at low to moderate levels of N deposition and no or adverse effects

at high levels. In line with these findings from observational studies, experimental studies also report more pronounced reactions of soil solution and foliar concentrations to decreased nitrogen deposition compared to the other parameters. Stevens (2016) reviewed experimental and observational studies in grasslands, heathlands, wetlands, and forests for information on the speed of recovery from high N deposition. Mainly in line with our findings, they report a relatively rapid response for mobile or plant-available forms of N in soil chemistry and for N contents in plant tissues across habitats (with the exception of forests showing a slower response in foliar element concentrations compared to other habitats). Similarly, Rowe et al. (2017) suggest N leaching rates and (moss) tissue N concentrations as midpoint-metrics, i.e. as indicators for effects-based monitoring of progress towards pollution reduction targets, due to their dynamic response to changing N deposition rates.

Linking results from observational and experimental studies is problematic due to the more controlled conditions and the typically faster and stronger cutback of N supply rates in experimental settings compared to real-world decreases in N deposition. A multitude of confounding factors, including the joint decrease of N and S deposition (e.g. Armolaitis and Stakenas, 2001) complicate the interpretation of results from observational studies. Furthermore, many of the large-scale observational studies reviewed in this paper are based on plots which are not distributed representatively across Europe. The larger monitoring efforts in Central and Western Europe likely led to an overrepresentation of plots where N deposition remained on a high level despite comparatively large decreases of N deposition.

Future decrease of N deposition to forests in Europe and associated ecosystem responses will most likely be limited (Fig. 3). Simpson et al. (2014) expect only minor reductions in the European ecosystem area with exceedances of the critical load for nutrient nitrogen (from 64% in 2005 to 50% in 2050). Under the assumption that soil solution NO_3^- concentrations and potentially also foliar N concentrations track changes in N inputs with a delay of only a few years (see above), limited changes of these parameters in response to declining N deposition would be expected for the future. For tree vitality and vitality-related growth effects, time-lags in the recovery from excess N deposition might be expected due to slow reversal of N-induced soil acidification and changes in mycorrhizal association. For understory vegetation community composition it has to be questioned whether full recovery can be expected at all since forest biodiversity is facing a number of additional “extinction debts” such as habitat loss and fragmentation, climate impact, and non-native species invasion (see e.g. Perring et al., 2017) likely causing further decline in biodiversity (Essi et al., 2015). If at all, these recovery processes will, however, only become apparent in regions with sufficient absolute magnitude of the cutback in N deposition. Furthermore, responses will likely be highly heterogeneous in space controlled by site-specific conditions.

In view of our results, a simple reversal of the stages of the classical nitrogen saturation concept (Fig. 1) does not seem to reflect the observed and expected responses to decreasing N deposition appropriately. Instead, several forest ecosystem properties seem to react with varying degrees of delay to cutbacks in N deposition. Correspondingly, the overall forest ecosystem state develops on a different trajectory during the process of N desaturation compared to N saturation. This hysteresis behavior is in line with findings from Gilliam et al. (2018, in press), who review results for soil acidification, plant biodiversity, soil microbial communities, forest carbon (C) and N cycling, and surface water chemistry focusing on the US. In view of the high variability of forest ecosystems, a set of “recovery types” could potentially serve to roughly classify the development of major strata of forest sites

under decreasing N deposition. For analytic and predictive purposes, more detailed models will be required to adequately represent processes of N (de-)saturation. In particular, dynamic modelling approaches taking complex microbial soil N processes into account may provide insights into the developments of forest ecosystem N pools accumulated over the last decades (Akselsson et al., 2016; Bonten et al., 2016; Dirnböck et al., 2017; Fleck et al., 2017; Rizzetto et al., 2016; Yu et al., 2016). Under the expected limited future decrease in N deposition, other controlling factors like climate change and forest management strategies will probably dominate the changes in N-enriched forests.

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Declarations of interest

None.

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